

AMENDMENTS TO THE CLAIMS

This listing of claims will replace all prior versions, and listings, of claims in the application.

Listing of Claims:

1-14 (Canceled)

15. (New) A method of production of an organoalkoxydialkylsilane of formula:

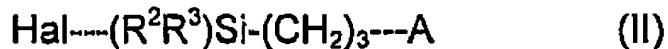


comprising the steps of:

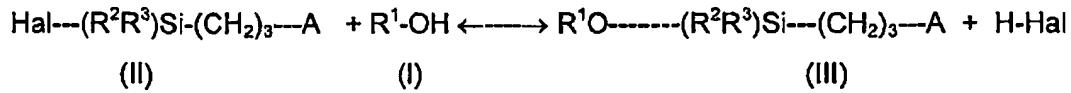
a) bringing an alcohol of formula:



into contact with a silane of formula:



in order to carry out the reaction of alcoholysis of said silane according to the following equilibrium reaction:



wherein:

the symbol Hal represents chlorine, bromine or iodine atoms,

the symbols R¹, which are identical or different, each represent a monovalent hydrocarbon group being a linear or branched alkyl radical having from 1 to 15 carbon atoms and a linear or a branched alkoxyalkyl radical having from 2 to 8 carbon atoms;

the symbols R² and R³, which are identical or different, each represent a monovalent hydrocarbon group being a linear or branched alkyl radical having from 1 to 6 carbon atoms or a phenyl radical;

A represents a detachable being a chlorine, bromine or iodine atom; a para-R⁰-C₆H₄-SO₂-O- radical wherein R⁰ is a linear or branched C₁-C₄ alkyl radical; a R⁰-SO₂-O- radical wherein R⁰ is as defined above; or an R⁰-CO-O- radical wherein R⁰ is as defined above;

b) working, under a pressure equal to or different from atmospheric pressure, in a stirred reactor equipped with a distillation column and with a condenser with possibility of reflux;

c) carrying out, on the one hand, the said bringing into contact of step a) by pouring the alcohol of formula (I) into a mixture of silane of formula (II) + organic solvent(s) phase, it being possible for the solvent(s) phase to be used partly, mixed with the alcohol of formula (I), and on the other hand the removal of the halogenated acid formed of formula H-Hal by entrainment by means of said organic solvent(s) phase, and

d) recovering the organoalkoxydialkylsilane of formula (III) formed in the reactor said process being characterized by the following points:

the mixture of silane of formula (II) + organic solvent(s) phase is heated to a temperature that corresponds to its boiling point in the pressure conditions prevailing during execution of the process, and introduction of the alcohol begins when the condenser on the reactor is charged and is operating in conditions of steady-state reflux;

the solvent(s) phase comprises one or more organic solvents selected so as to be able to fulfill a dual function: on the one hand, remove the halogenated acid formed by entrainment and salting-out of the gas owing to very low affinity of said phase for the acid, and on the other hand to provide liquid-vapour equilibrium with the alcohol which provides a concentration of alcohol of formula (I) in the mixture of alcohol of formula (I) + organic solvent(s) phase in the range from 5 to 30 wt.%;

the manner of introduction of the alcohol of formula (I) follows an operating procedure that is designed to prevent, at any moment during the reaction of alcoholysis of the silane of formula (II), accumulation of the halogenated acid of formula H-Hal in the reactor by dissolution in the alcohol of formula (I), in such a way that the amount of halogenated acid entrained by the solvent(s) phase represents, at any moment during the reaction of alcoholysis of the silane of formula (II), more than 90 wt.% of the halogenated acid formed;

the total amount of alcohol of formula (I) introduced is such that the molar ratio of alcohol of formula (I) to silane of formula (II) is in the range from 1 to a value below 3; and

the amount of solvent(s) phase present in the reactor along with the silane of formula (II) depends on the nature of this phase and is determined so as to make it possible to limit, during introduction, the concentration of alcohol of formula (I) in the mixture of alcohol of formula (I) + organic solvent(s) phase in the aforesaid range from 5 to 30 wt.%.

16. (New) The method as claimed in claim 15, wherein the solvent(s) phase comprises solvent(s) whose boiling point, in the conditions of pressure prevailing during execution of the method, is close to that of the alcohol of formula (I).

17. (New) The method as claimed in claim 15, wherein the manner of introduction of the alcohol of formula (I) is based on operating procedure VA1, comprising:

at least two charges of two fractions of alcohol, the first fraction of alcohol corresponding to a proportion representing 60 to 90 mol.% relative to the total molar quantity of alcohol used,

at least two periods of reflux without charging, each of them subsequent to each alcohol charge effected; and

the flow rate and the charging time of each fraction of alcohol as well as the duration of each period of reflux without charging being controlled in such a way that each fraction of alcohol charged is

consumed during the period of reflux without charging that follows said charging.

18. (New) The method as claimed in claim 15, wherein the manner of introduction of the alcohol of formula (I) is based on operating procedure VA2, which comprises execution of a single continuous charge of alcohol, but employing a flow rate that decreases with the degree of progress of the reaction of alcoholysis of the silane of formula (II) in such a way that the rate of introduction of the alcohol tracks its rate of consumption, and this single charging stage can be extended by a period of reflux without charging of variable duration.
19. (New) The method as claimed in claim 15, wherein it employs an anhydrous alcohol containing less than 1000 ppm of water and the total quantity of alcohol of formula (I) introduced is such that the molar ratio of alcohol of formula (I) / silane of formula (II) is in the range from 1.05 to 2.5.
20. (New) The method as claimed in claim 15, wherein it employs a quantity of solvent(s) phase that is determined so as to provide a concentration of alcohol of formula (I) in the combination of alcohol of formula (I) + solvent(s) phase that is in the range from 10 to 30 wt.%.
21. (New) The method as claimed in claim 15, wherein the method is carried out with reactants of formulas (I) and (II) wherein:
the symbol Hal is a chlorine, bromine or iodine atom;
the symbols R¹ are, ethyl, n-propyl, isopropyl, n-butyl, CH₃OCH₂-, CH₃OCH₂CH₂- or CH₃OCH(CH₃)CH₂- radicals; and

the symbols R² and R³ are methyl, ethyl, n-propyl, isopropyl, n-butyl, n-hexyl or phenyl.

22. (New) The method as claimed in claim 21, wherein R¹ is methyl, ethyl, n-propyl or isopropyl radicals, and the solvent or solvents is/are hexane, heptane, or cyclohexane used alone or mixed with pentane.

23. (New) The method as claimed in claim 15, wherein, at the end of the alcoholysis reaction, distillation of the reaction mixture is carried out in order to remove the unconsumed alcohol and the solvent(s) phase which are optionally recycled to a new reaction of alcoholysis.

24. (New) The method as claimed in claim 23, wherein, the unconsumed alcohol and the solvent(s) phase are recycled to a new reaction of alcoholysis and the following sequence of operations is carried out:

introduction of the distillate based on alcohol and solvent(s) obtained from a previous operation, into the reactor containing a new charge of silane of formula (II), optionally adding fresh alcohol and/or additional solvent(s) phase so that the concentration of alcohol of formula (I) in the mixture of alcohol of formula (I) + organic solvent(s) phase is within the range from 5 to 30 wt.%; then,

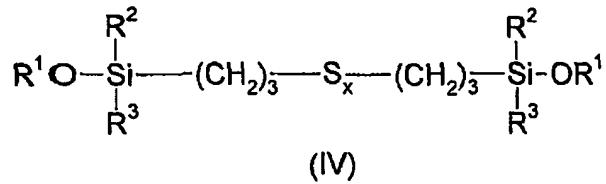
heating the mixture to raise its temperature to the value corresponding to its boiling point in the conditions of pressure prevailing during execution of the method and establishment of conditions with total steady-state reflux; then

carrying out a period of reflux without charging of alcohol during the time required for chemical consumption of the alcohol present in the reaction mixture; then

charging, either in batch mode or continuously, of the extra amount of alcohol required so that the molar ratio of total alcohol of formula (I) to silane of formula (II) is in the range from 1 to a value of less than 3; and, then

completion of the reaction by carrying out a second period of reflux without charging to reach a degree of transformation (TT) of the silane of formula (II) equal to at least 96 mol.%.

25. A method of production of the polysulfides of average general formula (IV):



wherein:

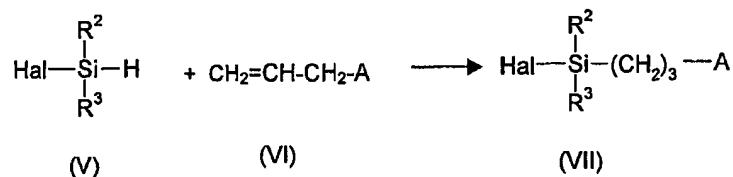
x is an integer or a fractional number, in the range from 1.5 ± 0.1 to 5 ± 0.1 ; and the symbols R¹, which are identical or different, each represent a monovalent

hydrocarbon group being a linear or branched alkyl radical having from 1 to 15 carbon atoms and a linear or a branched alkoxyalkyl radical having from 2 to 8 carbon atoms;

the symbols R² and R³, which are identical or different, each represent a monovalent hydrocarbon group being a linear or branched alkyl radical having from 1 to 6 carbon atoms or a phenyl radical;

said method being carried out as a sequence of stages (a), (b) and (c), in the definition of which the detachable group A corresponds to the symbol Hal representing a halogen atom and is a chlorine atom:

stage (a) corresponding to the process taking place according to the equation:



wherein:

the symbol Hal represents a chlorine atom, and

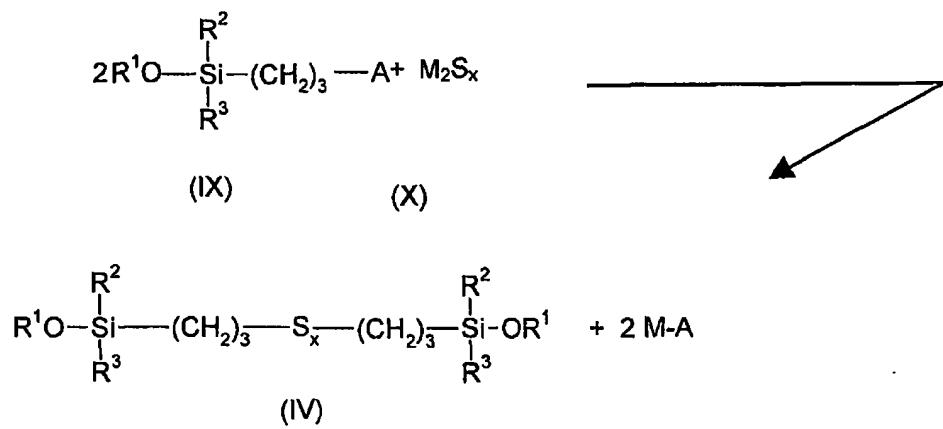
the symbols R² and R³ are as defined above, and A represents a detachable being a chlorine, bromine or iodine atom; a para-R⁰-C₆H₄-SO₂-O- radical wherein R⁰ is a linear or branched C₁-C₄ alkyl radical; a R⁰-SO₂-O- radical wherein R⁰ is as defined above; or an R⁰-CO-O- radical wherein R⁰ is as defined above;

the reaction being carried out:

by reacting, at a temperature in the range from -10°C to 200°C, one mole of the diorganohalosilane of formula (V) with a stoichiometric or non-stoichiometric molar quantity of the allyl derivative of formula (VI), working, in a homogeneous or heterogeneous medium, in the presence of an initiator comprising:

either a catalytic activator comprising: (i) at least one catalyst containing at least one transition metal or a derivative of said metal, selected from the group

consisting of Co, Ru, Rh, Pd, Ir and Pt; and optionally (2i) at least one hydrosilylation reaction promoter or auxiliary, or a photochemical activator, optionally comprising suitable ultraviolet radiation or suitable ionizing radiation, and optionally isolating the diorganohalosilylpropyl derivative of formula (VII) formed; stage (b) corresponding to the process described in claim 15; and stage (c) corresponding to the process employing the reaction:



wherein:

the symbols R^1 , R^2 , R^3 , A and x are as defined above, and

the symbol M represents an alkali metal,

the reaction being carried out:

by reacting, at a temperature in the range from 20°C to 120°C, either the reaction mixture obtained at the end of stage (b), or the monoorganooxydiorganosilylpropyl derivative of formula (IX) used separately after separation from said reaction mixture, with the metal polysulfide of formula (X) in the anhydrous state, using 0.5 ± 15 mol.%

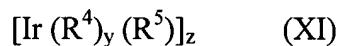
of metal polysulfide of formula (X) per mole of reactant of formula (IX) and optionally working in the presence of an inert polar (or nonpolar) organic solvent, and

- isolating the bis-(monoorganooxysilylpropyl) polysulfide of formula (I) formed.

26. (New) The method as claimed in claim 25, wherein stage (a) is carried out in the presence of an activator comprising, as the catalyst or catalysts (i), one and/or another of the following metallic species: (i-1) at least one finely-divided elemental transition metal; and/or (i-2) a colloid of at least one transition metal; and/or (i-3) an oxide of at least one transition metal; and/or (i-4) a salt derived from at least one transition metal and an inorganic carboxylic acid; and/or (i-5) a complex of at least one transition metal provided with organic ligand(s) that can possess one or more heteroatom(s) and/or with organosilicon ligand(s); and/or (i-6) a salt as defined above where the metallic part is provided with ligand(s) as also defined above; and/or (i-7) a metallic species selected from the aforementioned species (elemental transition metal, oxide, salt, complex, complexed salt) where the transition metal is associated in this case with at least one other metal selected from the family of the elements of groups 1b, 2b, 3a, 3b, 4a, 4b, 5a, 5b, 6b, 7b, and 8 (except Co, Ru, Rh, Pd, Ir and Pt) of the periodic table as published in the *Handbook of Chemistry and Physics*, 65th edition, 1984-1985, CRC Press, Inc., said other metal being used in its elemental form or in a molecular form, said association possibly giving rise to a bimetallic or multimetallic species; and/or (i-8) a metallic species selected from the aforementioned species (elemental transition metal and association of transition metal – other metal; oxide, salt, complex and complexed salt based on a transition metal or based on an

association of transition metal – other metal) which is supported on an inert solid support such as alumina, silica, carbon black, a clay, titanium dioxide, an aluminosilicate, a mixture of oxides of aluminum and zirconium, a polymeric resin.

27. (New) The method as claimed in claim 25, wherein stage (a) is carried out in the presence of an activator comprising:
as the catalyst or catalysts (i), at least one metallic species belonging to the iridium complexes of formula:



in which:

the symbol R^4 represents either a monodentate ligand L and in this case $y = 2$, or a bidentate ligand $(\text{L})_2$ and in this case $y = 1$, and
the symbol R^5 represents an halogen atom, and in this case $z = 2$, or a ligand of type LX and in this case $z = 1$;

and as the optional auxiliary or auxiliaries (2i), at least one species in the free state or supported, selected from the group of compounds consisting of:

- (i) the ketones,
- (ii) the ethers,
- (iii) the quinones,
- (iv) the anhydrides,
- (v) the unsaturated hydrocarbons (UHC) having an aromatic character and/or containing at least one $\text{C}=\text{C}$ double bond and/or at least one $\text{C}\equiv\text{C}$ triple bond, where these unsaturated bonds can be

conjugated or unconjugated, said UHCs being linear or cyclic (mono- or polycyclic), having from 4 to 30 carbon atoms, having from 1 to 8 ethylenic and/or acetylenic unsaturations and optionally containing one or more heteroatoms, and

(vi) and mixtures thereof,

with the condition that when the auxiliary comprises one or more UHC as defined above, this UHC or these UHCs is/are mixed with at least one other auxiliary different from a UHC.

28. (New) The method as claimed in claim 25, wherein stage (c) is carried out using anhydrous metal polysulfides of formula (X) which are prepared beforehand from an alkali metal sulfide M_2S in the form of a hydrated sulfide, by a process involving a sequence of the following steps (1) and (2):
step (1), involving dehydration of the hydrated alkali metal sulfide by applying a suitable method by which the water of crystallization can be removed while keeping the alkali metal sulfide in the solid state, throughout the dehydration step;
step (2), in which one mole of dehydrated alkali metal sulfide obtained is then brought into contact with $n(x-1)$ moles of elemental sulfur, working at a temperature in the range from 20°C to 120°C, optionally under pressure and also optionally in the presence of an anhydrous organic solvent, the aforementioned factor n being in the range from 0.8 to 1.2 and the symbol x being as defined above.